The Effect of Gas Pressure and Gas Flow Rate on the Magnetic Properties of Sputtered Ni and $Ni_{81}Fe_{19}$ Films

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***Abstract*—Magnetic properties of Ni and $Ni_{81}Fe_{19}$ films prepared by magnetron sputtering under different gas pressures and flow rates with a thickness of ~ 40 nm were investigated using magneto-optical Kerr effect magnetometry. Wavelength-dispersive X-ray analysis was used also to ascertain the formation of these films. These measurements confirmed that these films were free from contamination of argon atoms/ions. For Ni–Fe films prepared under low gas flow rates, the Ni weight ratios at all gas pressures were found to be higher than the films which were prepared under high gas flow rates and vice versa for the Fe weight ratios. A slight variation in the weight ratios of Ni and Fe across the Ni–Fe films area was noticed. In both Ni and Ni–Fe samples prepared under >6 mT, the coercivity was sharply increased by increasing the gas pressure and gas flow rate. These variations were attributed to the increment of the number of collisions of atoms with each other and/or with the gas particles/ions and changes in both the magnetization behavior and their elemental composition. This article suggested low gas pressure to be used during deposition to be used in magnetic devices.**

***Index Terms*—Ferromagnetic thin films, Gas flow rates, Gas pressure, Magnetic properties, Magnetron sputtering.**

I. Introduction

Magnetic thin films of nickel, cobalt, iron, and their alloys are showing an interest from industrial points of view and for exploring the fundamental physics. For example, they have many technological applications, in particular, giant magnetoresistance, magneto-optical devices, and ultra-high-density magnetic storage media in hard disc drives, magnetic random-access memory, and magnetic sensors [1-5].

A wide range of deposition techniques has been utilized to fabricate these films, which include chemical vapor deposition (CVD) [6,7], electroless plating [8], and electrodeposition [9-11]. Each technique has, however, its own advantages and disadvantages. Sputtering is one of these techniques and it is one of the most important methods used in CVD techniques. Sputtering is defined as the removal of surface material when it is bombarded with highly energetic ions and it has been proved to be simple, fast, and low-cost technique. This technique was first explored in 1852 by W. R. Grove when he was working on the electrical conductivity of gases [12,13]. In a sputtering technique, an electric field is applied between the cathode and the anode electrodes that are sited in an evacuated chamber. These electrodes can be connected to a D.C. or R.F power supply [12]. The sputtering chamber is filled with a low-pressure inert gas such as argon. This gas becomes positively ionized on applying a sufficient electric field to strike the glow discharge (plasma) between the anode and the cathode electrodes [12,13]. Due to the negative voltage of the target, the positive ions are accelerated toward the target. As a result, different phenomena can occur depending on the type of the material making up the target, the ion type, the ion energy, and other factors including gas flow rate, gas pressure, gas type, plasma power, electrode dimensions, electrode spacing, and the ratio of the electrode dimensions to the electrode spacing [6,7,12,13]. These phenomena are the ejection of atoms from the target, scattering and neutralization of ions, the production of the secondary electrons and ions, and ion implantation in the target with or without simultaneous target atom ejections.

The crystalline structure and the magnetization behavior of a range of ferromagnetic thin films which were produced by this technique were investigated extensively over the past few decades by changing the parameters or the conditions under which they were prepared [14-19]. For instance, type of the inert gas used [14], target history and geometry [15], separation distance between the electrodes, the deposition plasma power, and the deposition rate and mode (AC or DC), as well as, film thickness, annealing, and the effect of the stray magnetic field from the target on the film magnetic properties were analyzed using different techniques including transmission electron microscopy with selected area electron diffraction and X-ray diffraction analysis. Furthermore, the effect of these parameters on their magnetization behavior was carried out using a range of characterization techniques [14-19], such as a superconducting quantum

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interference device, vibrating sample magnetometer, torque magnetometer, alternating gradient magnetometers, and magnetic/atomic force microscopy.

MOKE magnetometry [20-27] has not been widely used to study the magnetic properties of such ferromagnetic sputtered thin films. In this setup, however, the Kerr reflected signal is proportional to the amount of magnetization, which depends on the polarization rotation of a linearly polarized light following its reflection from a ferromagnetic medium [22]. This creates a very sensitive probe that is proportional to the change in the sample magnetic state to a depth of the order of the skin depth. This is due to the absorption of light in the medium which is called the optical skin depth. This parameter is very essential for probing the surface magnetic properties of such magnetic materials.

Very limited researches have been published discussing the effect of gas pressure and gas flow rates on the surface magnetic properties of such ferromagnetic samples using MOKE magnetometry. Therefore, the main purpose of this article is to investigate the effect of gas pressure and gas flow rates on the surface magnetic properties of Ni and $Ni_{81}Fe_{19}$ films created by DC magnetron sputtering system with fixing all other conditions and parameters using highly sensitive MOKE magnetometry.

II. Experimental Procedure

Here, ferromagnetic Ni and $Ni_{81}Fe_{19}$ films were deposited using the DC magnetron sputtering system. Before the deposition process, Si/SiO_2 substrates of an area of approximately 4 × 4 mm^2 were cleaned ultrasonically using acetone followed by isopropanol alcohol and dried with nitrogen gas. The lowest base pressure reaching within this system was ~2×10^{-6} T using a combination of rotary and turbomolecular pumps. This may have been limited by the very large O-ring seal on the top flange. The sputtering gas was argon which entered the vacuum chamber through a variable leak valve. By partially opening or closing this valve, the gas flow rate in the chamber can be controlled with low, medium, and high flow rates. The target material used in this investigation was Ni and $Ni_{81}Fe_{19}$ discs with a ~50 mm diameter and 99.99% purity. The distance between the anode and the target surface (cathode) was kept constant at ~3 mm. The distance between the target surface and the substrate holder was also kept constant at ~90 mm throughout the work presented here. The anode and the cathode were connected to a DC power supply, to provide the power needed to strike the argon plasma and maintain it. The target voltage and plasma current were measured with digital multimeters. The deposition rate was fixed to around 2–3 A°/s, and the film thickness was fixed to ~40 nm. The deposition rate and film thickness were monitored during the film growth using a quartz crystal oscillator. This oscillator was calibrated using grazing incidence X-ray reflectivity measurements [24,25]. The time required to deposit these films was around 170–219 s. The plasma power was fixed to be around 110–170 W. The variation in the plasma power depended on the gas pressure and gas flow rate in the sputtering chamber. The pressure used here to deposit such materials was approximately 3-8 mT under two different (low and high) gas flow rates.

A special sample holder was designed in this article to deposit multiple thin films at different gas pressures and gas flow rates within the same vacuum deposition run. This multiple substrate holder allows the deposition of up to 15 samples in a single run without breaking the vacuum. The maximum stray magnetic field measured near the sample holder was around 20 Oe.

To obtain a uniform film thickness across the whole sample area, the substrate holder was rotated during the deposition process by an electrical motor at ~12 revolutions per minute. This was selected according to successive experiments. Before the deposition, the chamber was flushed with argon gas for approximately 5 min, to remove the residual air molecules or water vapor inside the chamber if any were left after evacuation. To clean the target surface from oxides or any other contaminants, a pre-sputtering process was carried out before the film deposition for a time of ~5 min, while the sample shutter was closed.

To ascertain the elemental composition of the deposited Ni and $Ni_{81}Fe_{19}$ films and to calculate the weight ratios of Ni and Fe in the Ni–Fe films, wavelength-dispersive X-ray (WDS) analysis in scanning electron microscopy SEM Hitachi-SU 70 system was utilized with the support from [28].

The magnetic properties investigated in this article were performed at many different locations on the samples which prepared under different gas pressure and gas flow rate using a highly sensitive MOKE magnetometry in the longitudinal configuration at room temperature. In this system, a polarized laser light was focused into a spot of a diameter of ~5 μm. The variation in the polarization angle of the reflected laser light from these films was proportional to the longitudinal component of the sample surface magnetization into a depth of a few nanometers. An AC electromagnet with a maximum field of approximately ±450 Oe was used to switch the magnetization of such films at a frequency of around 21 Hz. The obtained hysteresis loops are the Kerr voltage against the external applied magnetic field. All the magnetic measurements were performed for repeated loops averaged over ~3 min.

To gain a full understanding of the magnetic distribution across the whole surface area of these films, the coercivity distribution and the angular dependence of the coercivity were carried out by applying the magnetic field at different angles with respect to an arbitrary sample long axis. More theoretical and experimental discussions on the MOKE setup can be found in the study of Maruyama, *et al*. [20] Allwood, *et al*. [21] Brundle, *et al*. [22] Gasche, *et al*. [23] Sultan, [24] Sultan, [25] Musaab, [26] David, [27].

III. Results and Discussion

Fig. 1a shows an example of WDS measurements of the sputtered $Ni_{81}Fe_{19}$ film, which was prepared under 7 mT and high gas flow rates. The atomic identification of the peaks in the spectrum is related to Ni–Fe, which reflect the actual composition of Ni–Fe film. Importantly, these analyses

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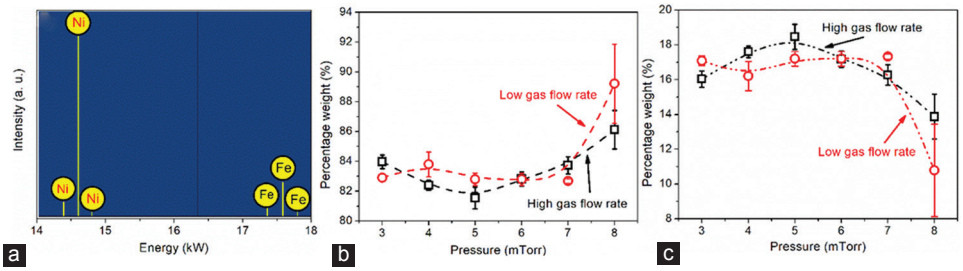

Fig. 1. (a) An example of wavelength-dispersive X-ray measurements of Ni81Fe19 thin film prepared under 7 mT and high gas flow rates by DC magnetron sputtering system. Elemental weight distributions of (b) Ni and (c) Fe in Ni–Fe films under different gas pressures and gas flow rates. The dashed lines are used as a guide to the eye and the error bars represent the elements distribution across the sample area.

confirm that these films are free from contamination of argon atoms or their ions seen in other investigation. Detailed WDS compositional analysis of Ni and Fe weight ratios of these films was undertaken for all samples which were prepared under different gas pressures and gas flow rates. The results of this work are plotted in Fig. 1b and c. Where, Fig. 1b and c shows the percentage of (b) Ni and (c) Fe weight ratios as a function of gas pressures for both low and high gas flow rates. These analyses demonstrate a remarkable effect of the gas pressure and gas flow rates on the percentage weight ratios of Ni and Fe in such films.

For all gas pressures investigated here, the percentage of Ni weight ratios prepared under low gas flow rates was slightly higher than their counterpart films which prepared under high gas flow rates and vice versa for the percentage of Fe weight ratios. There is clearly a slight variation of the elemental composition across the measured Ni–Fe film area up to ~3%, which has increased significantly up to ~6% by increasing the gas pressure and decreasing the gas flow rate, as demonstrated in the error bars shown in Fig. 1b and c. There is also a limited variation in the elemental weight ratios of Ni and Fe by increasing the gas pressure up to an approximately 5%. These elemental composition distributions might be due to the enhancement of the number of collisions of the ejected Ni and Fe atoms with each other and/or with other atoms or particles/ions exist in the sputtering chamber which may in turn reduce or increase one material than the other as a result of changing the gas pressure or gas flow rate. The differences in the size and the atomic weight of Ni and Fe materials might also contribute to these compositional discrepancies. This compositional modification, nonetheless, is likely to have some effect on the magnetic properties of these films grown under such conditions as will be seen in the succeeding discussion.

The magnetic properties of Ni and Ni–Fe films prepared under various gas pressures and gas flow rates were investigated here. These measurements were conducted by focusing the MOKE laser spot at many different positions on each sample when the magnetic field was applied at different angles with respect to an arbitrary sample long in-plane axes. There were no significant variations in the shape of the hysteresis loops obtained from different locations and different orientations with respect to the field applied when the films were prepared under <6 mT. This means that these films have no magnetic preferred orientation. The literature supports the isotropic nature of these films, whereas a large variation in the shape and size of the hysteresis loops was obtained at higher gas pressures (>6 mT) and lower gas flow rates.

To understand thoroughly the effect of gas pressure and gas flow rate on the magnetic properties of such films, the extracted coercivity from MOKE loops was plotted as a function of the gas pressure and gas flow rate as shown in Fig. 2. The error bars shown in the figure represent the distribution of coercivity across the sample area which was obtained by repeating MOKE measurements at different locations on the same sample. The coercivity distribution across the sample area might be attributed to the variations in the film properties, in particular, the surface defects or film non-uniformity which is discussed elsewhere [19,29]. The existence of such surface structural variations can act as pinning centers during the domain wall reversal and hence can change the coercivity and the shape of the hysteresis loop. Moreover, the differences in the elemental weight ratios of Ni–Fe films which have been discussed earlier are also likely to have some effect on the magnetic properties of such films.

For all gas pressures and gas flow rates, the coercivity of Ni films, as expected, is higher than the coercivity of Ni–Fe films. For instance, at a gas pressure <~6 mT, the average coercivity of Ni films is around 13 Oe, whereas for Ni Fe films is around 3 Oe. This result indicates that Ni films are slightly harder than Ni–Fe films. For the samples prepared under <~6 mT, the coercivity of both Ni and Ni–Fe films was found to be almost independent of the gas pressure and gas flow rate. Whereas, for the films deposited under more than ~6 mT, the coercivity of both the films was sharply increased by increasing the sputtering gas pressure up to ~8 mT. As an example, the average coercivity of the Ni and Ni_{81}–Fe_{19} films at a gas pressure of ~4 mT was found to be ~13 Oe and ~3 Oe, respectively, whereas, it significantly jumped to ~110 Oe and ~55 Oe, for Ni and Ni–Fe films, respectively, at a higher gas pressure of ~8 mT and low gas flow rate. Similar trends were noticed in other researches using other ferromagnetic layers [14,16]. The differences in the coercivity by changing the gas pressure and gas flow rate could be explained as follows.

At low gas pressure of <~6 mT, the ejected Ni and Ni–Fe atoms arrive the substrate with high thermal energies. These energies allow the atoms to have a high surface mobility at

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the Si/SiO_2 substrate. As the gas pressure increased in the sputtering chamber, the thermal energy of the atoms and their directions will be changed due to the enhancement of the number of collisions of these atoms with each other and/or with other atoms or particles/ions that exist in the sputtering chamber. Accordingly, the ejected Ni and Ni–Fe atoms will have low thermal energy at the substrate surface, which may in turn create voids and defects in these films and hence lead to the deviations in both the magnetic properties and the elemental composition variations in the alloy films. To confirm this explanation, the effect of gas pressure and gas flow rate on the deposition rate on the Si/SiO_2 substrate for both Ni and Ni–Fe materials is plotted in Fig. 3. Clearly, the deposition rate decreases with increasing the gas pressure in the sputtering chamber. Thus, the MOKE measurements suggest that the magnetic properties of such ferromagnetic films prepared under such conditions are strongly dependent on the gas pressure and gas flow rates in the chamber when they are more than 6 mT. Therefore, low gas pressure during the fabrication process of such ferromagnetic thin films is highly recommended to be used in magnetic devices.

IV. Conclusions

The effect of argon gas pressure and flow rate on the magnetic properties of Ni and $Ni_{81}Fe_{19}$ films prepared by DC magnetron sputtering system was investigated here in detail. The thickness of all these films was fixed to ~40 nm. WDS analysis confirmed the formation of these films without contamination of argon atoms/ions and other impurities. There was a remarkable effect of gas pressure and gas flow rate on the percentage weight ratios of Ni and Fe in the Ni–Fe films. There was a slight variation in the elemental composition across the measured Ni–Fe sample area up to ~3%, which increased significantly up to ~6% by increasing the gas pressure and decreasing the gas flow rate. There was also a limited variation in the elemental weight ratio up to an approximately 5% by increasing the gas pressure in the sputtering chamber.

There was no significant variation in the shape of the MOKE hysteresis loops obtained at different locations on these samples and different orientations with respect to field applied when the films were grown under <6 mT. Whereas, a large variation in the shape and the size of these loops were noticed at higher gas pressures (>6 mT) and gas flow rates. These variations in the elemental weight ratios of Ni–Fe films as well as the MOKE results were attributed to the increased collisions of the ejected atoms with each other and/or with other particles/ions that exist in the chamber which may in turn create defects or/and change the weight ratios in these films.

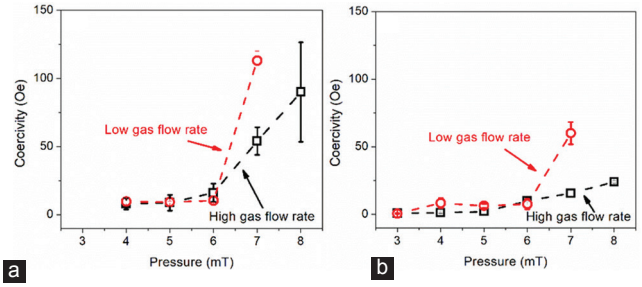

Fig. 2. The effect of gas pressure and gas flow rate on the coercivity of (a) Ni, and (b) Ni–Fe films. The error bars shown in the figures represent the variation in the coercivity across the sample area. The dashed lines are used as a guide to the eye.

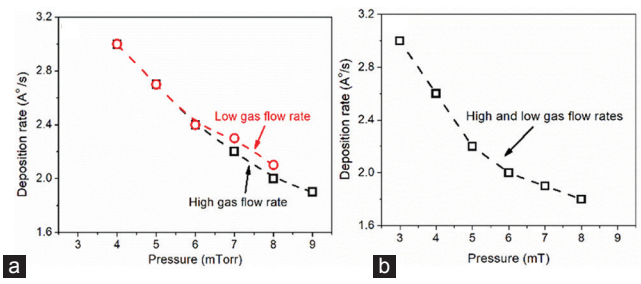

Fig. 3. Effect of gas pressure and gas flow rate on the deposition rate of (a) Ni and (b) Ni–Fe materials. The dashed lines are used as a guide to the eye.

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